

Charge dynamics in the normal state of the iron oxypnictide superconductor LaFePO

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We present the first infrared and optical study in the normal state of *ab*-plane oriented single crystals of the iron-oxypnictide superconductor LaFePO. We find that this material is a low carrier density metal with a moderate level of correlations and exhibits signatures of electron-boson coupling. The data is consistent with the presence of coherent quasiparticles in LaFePO.

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The recent discovery of superconductivity in the iron oxy-pnictides promises to be an important milestone in the field of superconductivity [1, 2]. Here is a new class of quasi-two-dimensional materials with a layered structure and relatively high superconducting T_c values [2, 3] rivalling the doped superconducting cuprates. Electronic conduction is believed to occur in the iron-pnictogen layers [4], similar to the cuprates where the charge carriers are delocalized in the copper-oxygen planes. While maintaining the cuprates as the benchmark for judging the new family of iron-based superconductors, it is worthwhile to note that the high superconducting transition temperatures are not the only puzzling features of the cuprates. Their normal state properties are unconventional and include pseudogap features, strong-coupling effects, charge-spin self-organization, and bad metal behavior [5]. Superconductivity arises from the Cooper instability of this anomalous metallic state of the cuprates. It is therefore imperative to investigate and understand the normal state of the iron-oxypnictide superconductors, especially their in-plane properties. Access to in-plane properties without contamination by the inter-plane response is only possible by studying single crystals.

In this Letter, we report on the infrared and optical response in the normal state of single crystals of LaFePO, the first superconducting iron-oxypnictide to be discovered [1]. The *ab*-plane infrared data allow us to examine the dynamical properties of the charge carriers in the iron-pnictogen layers. We find that LaFePO is a low-carrier density, moderately correlated metal with well-defined quasiparticles. Moreover, we find evidence of electron-boson coupling in the optical quasiparticle self-energy, demonstrating that many-body effects cannot be neglected.

The LaFePO crystals were grown by a flux method whose details are given in Ref.6. Resistivity and magnetization measurements reveal a superconducting T_c of ≈ 6 K, with complete Meissner shielding in the superconducting state. The crystals are platelets, typically $0.5 \text{ mm} \times 0.5 \text{ mm} \times 0.05 \text{ mm}$ in size. The *ab*-plane reflectance was measured in the near-normal incidence geometry in a Bruker v66 Fourier Transform Infrared Spectrometer at frequencies between 100 cm^{-1} and 24000 cm^{-1} . The reflectance measurements were performed at the follow-

ing temperatures: 298 K, 250 K, 200 K, 150 K, 100 K, 50 K, and 10 K. We obtained the optical constants through fitting the reflectance data with Drude and Lorentzian oscillators, and Kramers-Kronig constrained variational dielectric functions as described in Ref.7. In addition, variable-angle spectroscopic ellipsometry was performed in the frequency range 5500 cm^{-1} and 25000 cm^{-1} which improves the accuracy of the extracted optical constants in this frequency range.

The temperature dependence of the *ab*-plane reflectance of LaFePO crystals is displayed in Fig.1. This material is a very good metal as attested by the high values of the reflectance ($\geq 95\%$) at low frequencies. Two prominent hump-like features between 5000 cm^{-1} and 15000 cm^{-1} are likely due to interband transitions. In Fig.2, we present the real part of the optical conductivity $\sigma_1(\omega)$ over a broad frequency range. There is a clear Drude-like feature at low frequencies followed by two prominent inter-band transitions centered at 7100 cm^{-1} and 13000 cm^{-1} . A weaker interband transition centered at 3700 cm^{-1} becomes more evident in the $\sigma_1(\omega)$ data obtained at $T = 10 \text{ K}$. On the basis of band-structure calculations, we assign these interband transitions to the bands formed by the $3d$ orbitals of Fe [8, 9, 10]. Interestingly, the Drude part of the conductivity that represents intra-band excitations is well-separated from the higher-lying inter-band transitions. Such a separation is not usually evident in strongly correlated metals, specifically in most cuprates [5]. The difficulty in separating the Drude part from the interband transitions in the cuprates arises from the strong incoherent contribution to the conductivity in the mid-infrared frequency range [5].

A plasma frequency ω_p of 14900 cm^{-1} for the delocalized charge carriers is obtained by integrating the low frequency part of $\sigma_1(\omega)$: $\omega_p^2 = 8 \int_0^{\omega_c} \sigma_1(\omega) d\omega$. A cut-off frequency $\omega_c = 3000 \text{ cm}^{-1}$ was chosen above which interband transitions begin to dominate the optical response of this system. The frequency-dependence of the spectral weight associated with $\sigma_1(\omega)$ is plotted in the inset of Fig.2 in two ways: as the effective number of charge carriers per formula unit $N_{eff}(\omega)$ participating in the optical transitions [5], and as an energy $K(\omega)$ [11].

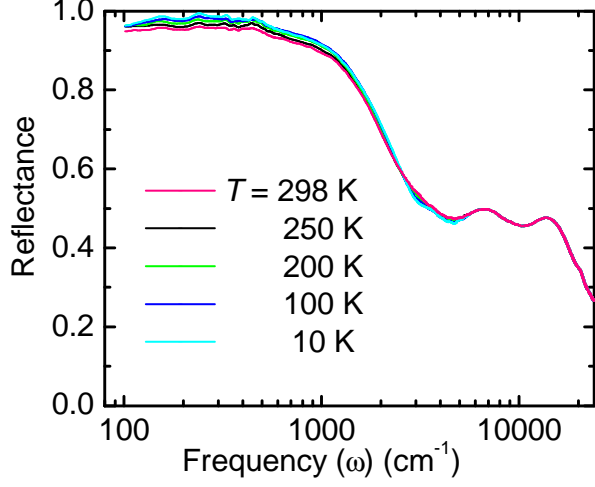


FIG. 1: (color online): Plots of *ab*-plane reflectance of single crystal LaFePO as a function of frequency for several representative temperatures.

$$N_{eff}(\omega) = \frac{2m_e V}{\pi e^2} \int_0^\omega \sigma_1(\omega') d\omega' \quad (1)$$

$$K(\omega) = \frac{\hbar c_0}{e^2} \int_0^\omega \frac{2\hbar}{\pi} \sigma_1(\omega') d\omega' \quad (2)$$

In the above equations, m_e is the bare electron mass, V is the volume of the unit cell per formula unit, and c_0 is the *c*-axis lattice constant. Assuming an intraband cutoff frequency of 3000 cm^{-1} , we estimate 0.16 itinerant charge carriers per formula unit. The energy $K(\omega)$ at the intraband cutoff $\omega_c = 3000 \text{ cm}^{-1}$ is simply the kinetic energy of the itinerant carriers and is equal to 0.15 eV. Correlations effects are expected to reduce the kinetic energy compared to the band theory value [11, 12]. Therefore, it would be useful to compare the experimental value with future band structure calculations for a quantitative estimate of the strength of correlations. In this context, one ought to mention that the kinetic energy in the cuprates near optimal doping is about 0.1 eV [12] and this is generally considered as evidence of strong correlations. The kinetic energy in LaFePO is higher than in optimally-doped cuprates, even though the effective number of itinerant carriers is similar [13], indicating lesser degree of correlations in LaFePO.

We plot the temperature dependence of the low-frequency part of $\sigma_1(\omega)$ in Fig.2b. The Drude-like peak at low frequencies becomes sharper and the conductivity at the lowest measured frequencies increases with decreasing temperature. The spectral weight is transferred from high frequencies to low frequencies with decreasing temperature, presumably below our low-frequency experimental cutoff. We also clearly observe an anomaly in $\sigma_1(\omega)$ near 500 cm^{-1} which becomes more prominent as the temperature is lowered. In order to gain

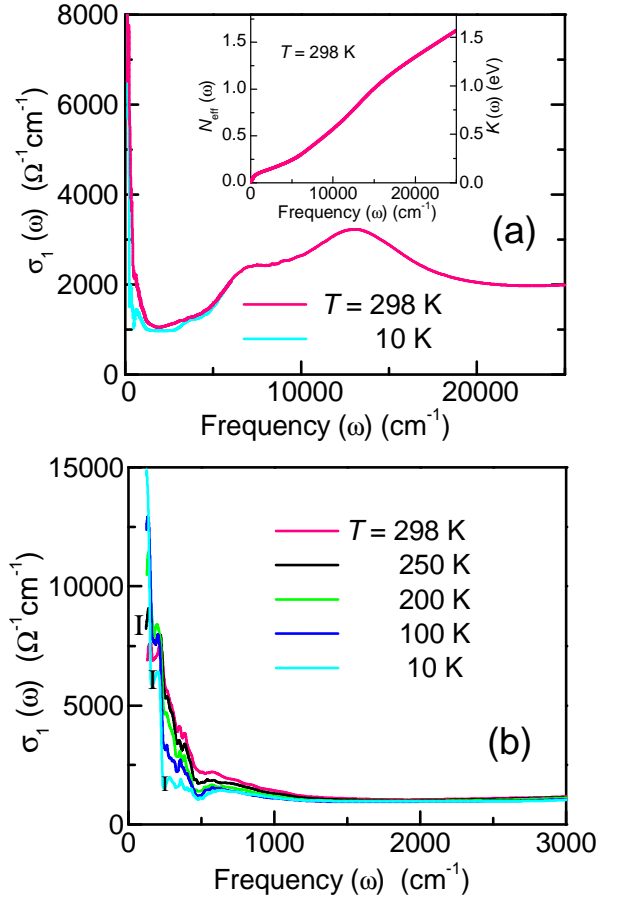


FIG. 2: (color online): (a) Real part of the *ab*-plane optical conductivity $\sigma_1(\omega)$ of LaFePO is plotted as a function of frequency for $T = 298 \text{ K}$ and $T = 10 \text{ K}$. The inset in panel (a) shows the frequency dependence of the effective number of charge carriers per formula unit $N_{eff}(\omega)$ and the energy $K(\omega)$ for $T = 298 \text{ K}$. (b) Systematic temperature dependence of $\sigma_1(\omega)$. The uncertainty in $\sigma_1(\omega)$ in panel (b) is less than or equal to the thickness of the lines for $\omega > 350 \text{ cm}^{-1}$. However, the uncertainty in $\sigma_1(\omega)$ increases at lower frequencies as indicated by the error bars.

more insight into this feature and to obtain the dynamical behavior of the charge carriers, we performed the extended Drude analysis [5, 14] on the real and imaginary parts of the optical conductivity $[\sigma_1(\omega), \sigma_2(\omega)]$. Via the extended Drude analysis, we extract the real and imaginary parts of the optical quasiparticle self-energy $[\Sigma_1^{op}(\omega), \Sigma_2^{op}(\omega)]$ which we present in Fig.3a,b. The effects of many-body interactions are encoded in $\Sigma_1^{op}(\omega), \Sigma_2^{op}(\omega)$. The self-energy is related to the optical conductivity, the scattering rate $1/\tau(\omega)$ and the mass enhancement factor $m^*(\omega)/m_b$ (m_b is the band mass) by the following equations:

$$-2\Sigma_2^{op}(\omega) = \frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi} \left(\frac{\sigma_1(\omega)}{\sigma_1^2(\omega) + \sigma_2^2(\omega)} \right) \quad (3)$$

$$1 - \frac{2}{\omega} \Sigma_1^{op}(\omega) = \frac{m^*(\omega)}{m_b} = \frac{\omega_p^2}{4\pi\omega} \left(\frac{\sigma_2(\omega)}{\sigma_1^2(\omega) + \sigma_2^2(\omega)} \right) \quad (4)$$

We note that in a system like LaFePO in which multiple bands cross the Fermi energy, the self-energy deduced from the infrared data is to be regarded as an average of the contributions from the relevant bands.

The anomaly in $\sigma_1(\omega)$ appears as a kink in the scattering rate at $\approx 500 \text{ cm}^{-1}$ for all measured temperatures between 298 K and 10 K. This kink has a weak temperature dependence - it becomes only marginally more pronounced at lower temperatures and the frequency of the onset of the upturn in $1/\tau(\omega)$ increases from 450 cm^{-1} at $T = 298 \text{ K}$ to 460 cm^{-1} at $T = 10 \text{ K}$. The kink in the scattering rate appears in the real part of the self-energy as a peak centered at $\approx 500 \text{ cm}^{-1}$ (62 meV) seen in Fig.3b. The most probable explanation of the kink in the scattering rate and the peak in real part of the self energy is in terms of the coupling of charge carriers to a bosonic mode [5, 14]. Infrared data alone cannot determine whether the bosonic mode is of phononic or magnetic origin. The kink in $1/\tau(\omega)$ may, alternatively, be due to a pseudogap [5]. However, this latter interpretation would be inconsistent with the recent Angle Resolved Photoemission Spectroscopy (ARPES) data on single crystals of LaFePO where the pseudogap is not seen [15]. The electron-boson coupling features identified in our infrared studies warrant further investigation by other spectroscopic probes, for example, ARPES with higher energy resolution, tunneling spectroscopy and neutron scattering. We note here that ARPES data on single crystals of the related iron-arsenide superconductor $(\text{Sr/Ba})_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ reveals kinks in the dispersion (energy range 15-50 meV) which appear to be related to the energy scales of magnetic excitations [16].

Above the kink feature, the scattering rate increases monotonically with frequency. Electron-phonon scattering theory predicts a constant, frequency-independent scattering rate above the cutoff of the phonon spectrum [5, 19]. Calculations of the phonon spectrum for LaFePO are not yet available. However, the cutoff is not likely to exceed 600 cm^{-1} inferred from calculations on the iron oxy-arsenides [17, 18]. Therefore, the absence of saturation of $1/\tau(\omega)$ in LaFePO suggests that in addition to electron-phonon scattering, there are other scattering channel(s) presumably arising from electronic correlations. The available experimental data on LaFePO suggests this material is close to a spin density wave (SDW) instability [20] although there is no evidence yet for an SDW ground state [21]. We note that partial nesting of the Fermi surface [15, 20] and/or coupling to magnetic excitations [16, 21] may explain the monotonic increase

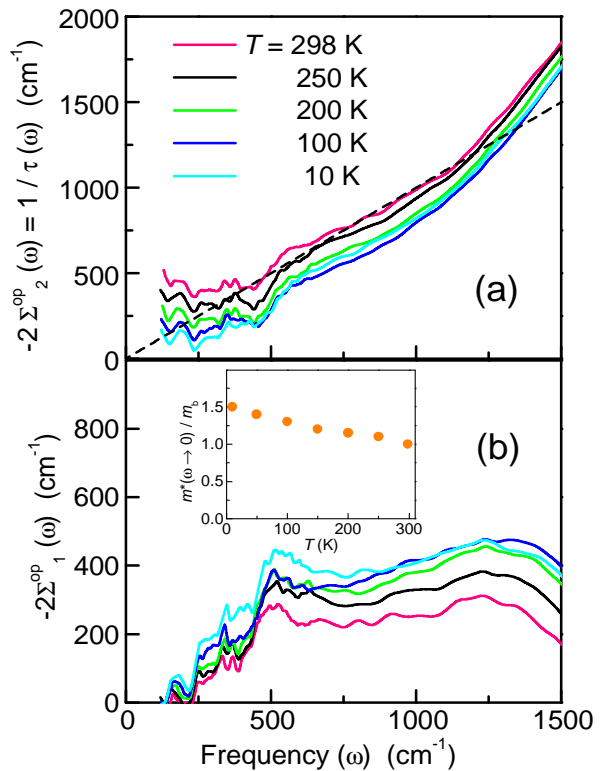


FIG. 3: (color online) Panels (a) and (b) respectively display the frequency dependence of the imaginary part of the optical quasiparticle self energy $-2\Sigma_2^{op}(\omega)$ [or scattering rate $1/\tau(\omega)$], and the real part of the optical quasiparticle self-energy $-2\Sigma_1^{op}(\omega)$ for representative temperatures in the normal state of LaFePO. The dashed line in panel (a) represents the equation $\omega\tau = 1$. The inset in panel (b) shows the temperature dependence of the mass enhancement factor in the low frequency limit $m^*(\omega \rightarrow 0)/m_b$.

of the frequency-dependent scattering rate of the quasiparticles. The scattering rate for LaFePO at the lowest measured temperature $T = 10 \text{ K}$ is close to or below the line $\omega\tau = 1$ at low frequencies (see Fig.3a). Within conventional Fermi Liquid Theory, the $\omega\tau = 1$ line separates well-defined quasiparticles from incoherent excitations [5, 19]. The data imply that well-defined quasiparticle excitations exist in LaFePO, consistent with the observation of deHaas-vanAlphen oscillations in all the Fermi surface pockets [20].

The mass enhancement factor ($\omega \rightarrow 0$) at $T = 10 \text{ K}$ is 1.5 ± 0.1 which is in reasonably good agreement with the factor of nearly 2 obtained from ARPES data and deHaas-vanAlphen measurements [15, 20]. The rather modest mass enhancement factor is only weakly dependent on temperature (see inset of Fig.3b) and these observations together suggest a moderate degree of electronic correlations.

It is instructive to compare the normal state infrared response of superconducting LaFePO to that of MgB_2

and the superconducting cuprates. First, the plasma frequency of LaFePO is comparable to the values seen in doped superconducting cuprates [5], indicating similar carrier densities in the two systems. This is in contrast to MgB₂, a high carrier density system, where the plasma frequency is nearly 48000 cm⁻¹ (≈ 6 eV) [22]. A linear frequency dependent scattering rate is seen in many of the cuprates near optimal doping and its origin still remains a mystery [5]. However, the $1/\tau(\omega)$ data in LaFePO does not show obvious signs of a linear frequency dependence. The scattering rate data extracted from infrared measurements on under-doped and optimally-doped cuprates lie above the $\omega\tau = 1$ line and suggests predominance of incoherent excitations [5] over a wide frequency range. This is to be contrasted with the predominance of quasiparticle excitations in LaFePO whose $1/\tau(\omega)$ data resembles what is seen in the over-doped cuprates which are considered closer to the Fermi Liquid regime [5]. In MgB₂, the scattering rate of charge carriers is significant, mainly due to strong electron-phonon coupling [22]. Hence, we infer that many-body interactions in LaFePO are weaker than in the under-doped and optimally-doped cuprates and MgB₂, but cannot be considered negligible.

The cuprates, especially low-doped ones, exhibit bad metal behavior [23] wherein the resistivity exceeds the Ioffe-Regel-Mott (IRM) limit of metallic transport and shows no signs of saturation at high temperatures. The IRM limit is generally expressed as $k_F l \approx 1$ (k_F is the Fermi wavevector and l is the electronic mean free path) or $l \approx a$ (a is the in-plane lattice constant). The IRM limit is regarded as a regime where the quasiparticle description is not valid [5, 19, 23]. At room temperature, the in-plane resistivity (ρ_{ab}) of LaFePO yields $k_F l \approx 2$ and this value increases by a factor of 25 just above T_c [6, 24]. At least at low temperatures, the $k_F l$ value is well above the IRM limit and therefore indicates that the quasiparticle description is applicable in LaFePO. In this context, it would be interesting to investigate the behavior of the resistivity of LaFePO at elevated temperatures.

The picture that emerges for LaFePO is of a low carrier density material with well-defined quasiparticle excitations leading to robust metallicity. Electronic correlations, though present, are weaker than in the optimally-doped cuprates. Moreover, there is evidence of electron-boson coupling in LaFePO. Hence, we classify LaFePO as a moderately correlated metal. It would be interesting to study the infrared properties of single crystals of the iron-arsenide compounds for comparison with our LaFePO results, especially the trends in the degree of electronic correlations.

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